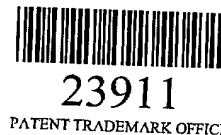


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September 15, 2000

Box PATENT APPLICATION

Assistant Commissioner for Patents Re: New U.S. Patent Appln.
Washington, D.C. 20231 Our Ref: 1748X/49133

Sir:

Transmitted herewith for filing is the patent application of:

Oskar Lamla and Martin Schüssler

entitled: **AN APPARATUS FOR A CARRYING OUT A HETEROGENEOUSLY
CATALYZED REACTION AND A METHOD FOR PRODUCING SAME**

Enclosed are:

1. Specification, including 11 claims and Abstract of the Disclosure (18 pages).
2. 2 Sheets of Formal drawings showing Figs. 1 - 4.
3. Declaration and Power of Attorney (**unexecuted**).
4. Information Disclosure Statement & Form PTO-1449 with 4 references.

Priority is being claimed under 35 U.S.C. §119 and 37 C.F.R. §1.55 based on Priority Document 199 44 185.5, filed in Germany on September 15, 1999.

The filing fee has been calculated as shown below:

Basic Fee						
Total Claims	<u>11</u>	-	20	=	<u>0</u>	x \$ 345/690 = \$690.00
Independent Claims	<u>4</u>	-	3	=	<u>1</u>	x \$ 9/18 = \$
						\$39/78 = \$ 78.00
Multiple Dependent Claim Presented						\$130/260 = \$
Total Filing Fee						<u>\$ 768.00</u>

The filing fee is being deferred.

Respectfully submitted,

Wan A 20/39085
Donald D. Evenson

for Registration No. 26,160

DDE:WAZ:msy



AN APPARATUS FOR A CARRYING OUT A HETEROGENEOUSLY
CATALYZED REACTION AND A METHOD FOR PRODUCING SAME

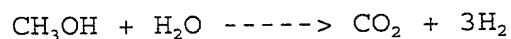
BACKGROUND AND SUMMARY OF INVENTION

This application claims the priority of German
patent document 199 44 185.5, filed 15 September 1999,
the disclosure of which is expressly incorporated by
reference herein.

The present invention relates to an apparatus for
carrying out a heterogeneously catalyzed reaction by
supplying a suitable reaction mixture to a catalyst, as
well as to a method for producing the apparatus.

An example of heterogeneously catalyzed reactions is
the production of hydrogen from a hydrocarbon or alcohol,
especially from methanol (i.e., methanol reforming). A
reaction mixture comprising a hydrocarbon or alcohol and
water is supplied to a catalyst. Further examples are
the carbon monoxide depletion with release of carbon
dioxide in a so-called hydrogen shift reaction; the
oxidation of carbon monoxide by supplying a gas-
containing carbon monoxide and a gas-containing oxygen to
a catalyst; as well as the combustion of a combustible
educt with addition of an oxygen-containing gas in a
catalytic burner.

The recovery of hydrogen from methanol is based on
the overall reaction



For carrying out this reaction, a reaction mixture
comprising a hydrocarbon and water vapor is passed along

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a suitable catalyst. Heat is supplied at the same time
in order to produce the desired hydrogen in a 2-step or
multi-step reaction. Such an apparatus for two-step
methanol reforming is known from EP 0 687 648 A1. In the
5 known apparatus, the reaction mixture is supplied to a
first reactor, in which a partial conversion of the
methanol occurs. After passing through the first
reactor, the gas mixture, in which portions of still
unreacted educt are contained, is passed into a second
10 reactor, which is optimized for the reaction. The
reactors are constructed as plate reactors or bulk
reactors, in which the catalyst is provided in the form
of a fill or a coating on the distributing channels.
Moreover, catalysts are known in the form of coated sheet
metal, nets and foams, through which the reaction mixture
flows.

From EP 0 217 532 B1, a method is known for the
catalytic production of hydrogen from mixtures of
methanol and oxygen using a gas-permeable catalyst
20 system, for which a hydrogen generator is provided with
an upper reaction zone and a lower reaction zone. The
reaction mixture of methanol and oxygen is supplied to
the upper reaction zone. After flowing through the upper
reaction zone, the reaction mixture is passed into the
25 lower reaction zone, in which, due to a spontaneous
initiation of the oxidation of the methanol, there is
such an increase in temperature, that a partial oxidation
of the methanol commences in the upper reaction zone in
30 the presence of a copper catalyst and hydrogen is formed.

EP 0 906 890 A1 discloses an apparatus for carrying
out a heterogeneously catalyzed reaction in which a

suitable reaction mixture is supplied to a catalyst, which is prepared by compressing a catalyst powder into a highly compressed layer forming a molded object. The reaction mixture can be forced through the catalyst layer with a drop in pressure. In order to seal them from the surroundings, such reactors usually have a massive metal housing.

It is an object of the present invention to provide an apparatus, which is an improvement with respect to weight and costs. Furthermore, it is an object of the present invention to provide a method for manufacturing such an apparatus.

This objective is accomplished by an apparatus and by a method according to the present invention.

The integration of a seal directly on or in the edge region of a stack of pressed layers represents a solution, which is an improvement with respect to weight and costs, since previously sealing of the edges was realized usually with a massive metallic housing. In contrast, pursuant to the present invention, a stack is sealed (1) in a stacking direction by end plates, and (2) transversely to the stacking direction by an integrated edge seal.

To increase safety, a sheet can be provided that surrounds the stack gas-tight. The escape of combustion gases through small leaks in the edge seal into the surroundings is avoided. For venting purposes, one or more openings provided with a catalyst can be provided. Accordingly, emerging combustion gases are reacted

completely with the oxygen of the surrounding air, so that contamination of the environment is reduced.

5 In an advantageous development of the present invention, the catalyst material is pressed together with a supporting structure, as a result of which the catalyst material is stabilized mechanically and/or the thermal conductivity is improved. The carrier structure advantageously is a three-dimensional network structure
10 (i.e., matrix) which, in a further advantageous development of the present invention, is a metallic supporting structure. Copper, especially dendritic copper, for example, is used as the metal.

15 For the preparation of the edge seal by an increased consolidation of the catalyst material in an edge region, it is only necessary to adapt the pressing tools and molds correspondingly. The tightness can be improved by admixing additional materials in the edge region, since
20 these materials can be selected especially with regard to their sealing function. Although a subsequent sintering step can represent an increased expense for the manufacture, this method of manufacturing is used preferably in cases in which the catalyst material is
25 also subjected to a sintering step, so that no additional effort is required.

30 In relation to quality control, the selection of a material that forms an edge seal under processing conditions is disadvantageous, since the tightness is brought about only after the equipment is started up. On the other hand, however, an additional manufacturing step can be omitted as a result.

A plurality of proven methods is available for applying a material, which is impermeable to gases and temperature resistant, on the finished stack of layers. The use of a soldering foil for sealing the edge has had very good results with respect to tightness.

Other objects, advantages and novel features of the present invention will become apparent from the following detailed description of the invention when considered in conjunction with the accompanying drawings.

BRIEF DESCRIPTION OF THE DRAWINGS

Figure 1 in a diagrammatic representation, shows the manner in which the inventive catalyst layer functions;

Figure 2 in perspective representation, shows an inventive stack-shaped arrangement of catalyst layers arranged in parallel;

Figure 3 shows a perspective representation of a further example of a single, inventive catalyst layer; and

Figure 4 shows a highly diagrammatic representation of a further example of an inventive stack-shaped arrangement of parallel catalyst layers in section.

DETAILED DESCRIPTION OF THE DRAWINGS

Figure 1 diagrammatically shows a side view of an inventive catalyst layer 10, which is formed by compressing catalyst material into a thin, highly compressed layer of large surface area. The layer 10

forms a molded object with a thickness d , which is, for example, 1 mm. As catalyst material, a finely grained catalyst powder or granulate is used, the particles of which have a diameter of 0.5 mm or less. The pressing takes place at temperatures, for example, of about 200°C to 500°C.

The catalyst layer 10, which is shown, is a component of an apparatus for producing hydrogen, the details of which are not shown. The educts of the reaction mixture, under the action of pressure, are supplied essentially perpendicularly to the catalyst layer 10 and pressed through this layer. On flowing through the catalyst layer 10, the reaction mixture experiences a pressure drop Δp of about 100 mbar and more (for example, 1 to 4 bar). On the opposite side of the catalyst layer 10, the catalytic reaction products emerge in the direction of the arrow.

An essentially gas-tight region is provided in the edge regions of the catalyst layer 10. This gas-tight region represents an edge seal 30, which ensures that the gas flows through the catalyst layer 10 only in the direction of the arrow and does not emerge laterally out of the catalyst layer 10. Due to the construction of an edge seal 30 directly in or on the catalyst layer 10, it is possible to do without a separate massive metal housing. The production of such an edge seal 30 is described in detail below.

In order to improve the mechanical stability and/or the thermal conductivity of the catalyst material, it is pressed into a support structure. This support structure

is a net-like matrix, which is obtained by mixing the at least one catalyst powder with dendritic copper in powder form and compressing this mixture. During the compression, the dendritic copper powder forms a net-like matrix structure, in which the catalyst particles are "incorporated". The dendritic copper powder, even when present in a relatively small amount in proportion to the total weight of the layer, can be compressed and/or sintered easily into a network, has a large surface area, and is itself catalytically active. Therefore, by using dendritic copper powder, a stabilizing, fixing and heat-distributing network in the micrometer range is obtained.

The catalyst layer 10 has a relatively large area of, for example, 100 cm². In order to achieve a compact construction, the catalyst volume, through which the reaction mixture flows, is divided into several layers, which are not next to one another but disposed one behind the other and parallel to one another. Such an arrangement is shown in Figure 2 and shows a stack 20, comprising a plurality of catalyst layers 10, 10', arranged one upon the other. In order to illustrate the mode of action better, the layers at the top in the drawing are shown at a distance from one another.

At its outer periphery, extending perpendicularly to the direction of the stack, each catalyst layer 10 has an edge seal 30. In addition, the catalyst layers 10 have channels 12, 14, 14', 16 for conducting educts and products of the catalytic reaction.

In the example shown in Figure 2, educt channels 12, extending parallel to the longitudinal edges, are

provided in the catalyst layer. The educt channels 12 form conducting channels proceeding perpendicularly to the plane of the catalyst layer. The educt channels 12 of catalyst layers 10, 10', lying on top of one another, are disposed essentially congruently to one another and thus form a continuous guiding channel for the educts of the reaction mixture from the top of the stack 20 to the bottom. A specific reaction mixture, which depends on the use to which the stack arrangement is put, is passed through the educt channels 12. In the event that the stack arrangement is used as a hydrogen reactor, the reaction mixture comprises alcohol, especially methanol, as well as chemically bound hydrogen, advantageously in the form of water. In the event that the stack 20 is used in a so-called hydrogen shift reaction for decreasing the carbon monoxide content and forming carbon dioxide, the reaction mixture comprises carbon monoxide and hydrogen. In the event that the stack is used in the area of carbon monoxide oxidation, the reaction mixture comprises a carbon monoxide-containing gas as well as an oxygen-containing gas. When the stack 20 is used in a catalytic burner, the reaction mixture comprises a combustible educt as well as an oxygen-containing gas.

The educt channels 12 of each second catalyst layer 10 are connected with distribution channels 14, which extend essentially parallel to the superficial extent of the catalyst layer 10 and discharge at least a portion of the reaction mixture, which enters through the educt channels 12, into the interior of the catalyst layer 10.

After that, one part of the reaction gas mixture, entering through the educt channels 12 and passed through

the stack 20, is discharged in each second layer plane through the distribution channels 14 into the interior of the two adjoining catalyst layers 10, 10', as a result of which a parallel switching of the catalyst layers, disposed one above the other, is achieved.

In the example, shown in Figure 2, two spatially separate educt channels 12 are provided per catalyst layer 10, 10', as described. This can be used for the purpose of supplying different materials of the reaction mixture separately from one another, so that individual components of the reaction mixture come together only in the plane of the catalyst layer 10.

Advantageously, a catalyst layer with a channel structure, as shown in the example of Figure 3, is used for this purpose. The catalyst layer 21, shown in Figure 3, has educt channels 22a, 22b and product channels 26, which in their function, in principle, correspond to the educt and product channels 12 and 16 described in connection with Figure 2. Deviating from the catalyst layer shown in Figure 2, the two educt channels 22a, 22b, which are disposed spatially separated from one another, are not connected to one another by the distribution channels. Instead, the distribution channels 24a or 24b, starting out from the educt channels 22a, 22b, extend transversely over the catalyst layer 21 and end before they reach the opposite educt channel 22b or 22a. An arrangement of alternately interlocking channels is created, which can be used for supplying separately a "further" gas, which is required for the reaction or supports this reaction. If, in the example of the methanol reformer, a mixture of methanol and water is

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supplied through the one educt channel (for example, the
educt channel 22a), oxygen (e.g., air) can be supplied
through the corresponding other educt channel 22b. Over
the distribution channels 24a, 24b, which are assigned to
5 the respective educt channel, the materials supplied are
distributed in the catalyst layer 21 and come into
contact with one another only in the layer itself. By
these means, a particularly homogeneous and safe (e.g.,
danger of explosion) distribution and mixing of the
10 educts is achieved. Embodiments, other than those shown,
with only one educt channel or also with more than two
educt channels, are of course also possible.

Along the transverse edges of the catalyst layers
10, 10', product channels 16, constructed analogously to
the educt channels 12, are disposed and form guiding
channels extending essentially perpendicularly to the
superficial extent of each catalyst layer 10. In the
case of superimposed catalyst layers 10, the product
channels 16 are congruent with the product channels of
the catalyst layers 10, 10' above and below. The product
channels 16 of each second catalyst layer 10' are
connected with collecting channels 14', which collect the
reaction product emerging from the catalyst layers 10,
25 10' that are above and below and, in the transverse
direction, lead to product channels 16 by which the
reaction products are discharged through the stack 20.

30 In the embodiment shown of an inventive apparatus
for carrying out a heterogeneously catalyzed reaction,
such as the generation of hydrogen, the superimposed
layers 10, 10' of catalyst have alternating modes of
functioning. In the catalyst layers 10, the educts,

supplied by the educt channels 12, are distributed over distributing channels over the surface of the catalyst layer above and below, flowing through this catalyst layer essentially perpendicularly and under an appreciable pressure drop. In the respectively following catalyst layer 10', the products of the catalytic reaction are collected in collecting channels 14' and supplied to the product channels 16 for discharging the reaction products from the stack 20.

Of course, embodiments are also conceivable, in which each catalyst layer takes over the supplying, distributing, collecting and discharging of the educts and/or products. Such more complex catalyst layers can be produced, for example, by pressing and sintering powdery catalyst material on already sintered catalyst layers.

With that, catalyst layers are made available, which can be produced in a simple and compact manner and are suitable for use in hydrogen reactors for the catalytic generation of hydrogen; hydrogen shift steps for depleting carbon monoxide; carbon monoxide oxidizers; and catalytic burners. Due to this configuration of the catalyst, a modular construction is possible, for which only slight thermal losses and no large temperature gradients occur, as a result of which a reaction that is homogeneous over a large volume can take place. The total catalyst volume can be reached spatially with educts, which leads to clearly improved starting dynamics. Moreover, the danger of ignition, of the homogeneous combustion of methanol or of the oxyhydrogen gas reaction is avoided.

By selecting suitable process parameters (e.g., compression pressure, temperature, and/or nature of the starting materials, such as particle size distribution, porosity, and the like), the expert can produce an catalyst layer or catalyst layer arrangement, which is tailor-made to meet the respective requirements and optimized with respect to the layer sequence, heat distribution, flow relationships, and mechanical properties, such as the pressure drop and stability.

The edge seal 30 can be produced in different ways. It can be formed owing to the fact that the catalyst powder, during the production of the edge region, experiences a stronger consolidation and, with that, forms an essentially gas-tight edge seal 30. Moreover, one or more other materials can be admixed with the catalyst powder in the edge region and then, in a subsequent processing step, form the essentially gas-tight edge seal 30. Preferably, materials are used that form a tight layer in a subsequent sintering step. It is, however, also possible to use materials, which are converted during process conditions and therefore, when the apparatus is started up for the first time, form the essentially gas-tight edge seal 30.

Furthermore, it is also possible to apply the edge seal 30 on the edge region after the catalyst layers 10 are pressed and stacked on top of one another. For this purpose, all materials, which are impermeable to gases and stable at elevated temperatures, can be used. Preferably, copper or a ceramic material is used. The edge seal 30 can be applied, for example, by immersion or

by plasma spraying or flame spraying. It is also possible to apply a soldering foil on the finished stack, the soldering foil, under the action of heat, penetrating into the porous body and then sealing it.

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The application of an edge seal 30 directly in or on the edge region of the catalyst layers 10 has the advantage that it is possible to do without the massive metal housing, which is otherwise customarily used. Thus, costs as well as the weight of the apparatus can be reduced.

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An apparatus, optimized with regard to safety, is shown in section in Figure 4. As in Figure 2, a stack 20 of catalyst layers 10, 10' is provided here. This stack is disposed between two end plates 31, 31', supplying and discharging lines 32 for the media being provided in one or both end plates 31, 31'. In the example shown, four supplying or discharging lines 32 are drawn in the upper end plate 31. This is, however, only a diagrammatic representation. Of course, a different number of supplying and discharging lines 32 can also be provided. In addition, these need not all be disposed in the upper end plate 31. Instead, a portion or all of the supplying and discharging lines 32 can be provided in the lower end plate 31'.

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In the edge region, all catalyst layers 10, 10' have an edge seal 30, which is only indicated by broken lines for the sake of simplicity and through which the stack 20 is sealed from the environment in a direction transverse to the direction of the stack. In the stack direction, the sealing is accomplished by end plates 31, 31'. For

30

5 this reason, the catalyst layers 10, 10' must, of course,
also be connected gas-tight with the end plates 31, 31'.
Furthermore, the whole of the stack 20 is surrounded by a
gas-tight sheet 33, which is also connected gas-tight
with the two end plates 31, 31'. The sheet 33 has at
least one opening 35, which is provided with an oxidation
catalyst 34. Combustion gases, which have emerged from
the edge seals 30 through tiny leaks that may be present,
after being oxidized completely with the oxygen present
10 in the surrounding air, can be discharged to the
surroundings through this opening 35. When the apparatus
is used in fuel cell systems, especially carbon monoxide,
hydrogen, methanol or hydrocarbons, which are oxidized to
carbon dioxide and water at a catalyst, come into
consideration here. Of course, several openings 35 can
also be provided. It is also possible to dispose the
opening 35 in one of the end plates 31, 31'. Aside from
an emission to the surroundings, a return to any place in
the system is also possible. Finally, the whole of the
20 apparatus can also be surrounded by the sheet 33, so that
a seal between the sheet 33 and the end plates 31, 31'
can be omitted.

25 The foregoing disclosure has been set forth merely
to illustrate the invention and is not intended to be
limiting. Since modifications of the disclosed
embodiments incorporating the spirit and substance of the
invention may occur to persons skilled in the art, the
invention should be construed to include everything
30 within the scope of the appended claims and equivalents
thereof.

WHAT IS CLAIMED IS:

1. An apparatus for carrying out a heterogeneously catalyzed reaction, comprising:

5 a stack comprising a plurality of layers on top of one another, each layer comprising a catalyst material and having channels for conducting educts of a reaction mixture and reaction products;

10 a plurality of end plates that bound said stack in a stacking direction, wherein at least one end plate has supplying or discharging lines that are connected with said channels;

wherein said plurality of layers have an edge seal (30) in an edge region or on a surface extending transversely to the stacking direction.

2. The apparatus of Claim 1, further comprising a gas-tight sheet that surrounds the plurality of layers.

20 3. The apparatus of Claim 2, wherein the gas-tight sheet has at least one opening for venting and comprises a catalyst.

25 4. The apparatus of Claim 1, wherein the catalyst material comprises a metallic support structure.

5. The apparatus of Claim 4, wherein the metallic support structure comprises dendritic copper.

30 6. A method for producing an edge seal for a stack of a plurality of layers, each layer comprising a catalyst material, said method comprising:

mixing a catalyst material with at least one additional material to form a mixture;

compressing the mixture, thereby forming a layer comprising the catalyst material and a gas-tight edge seal comprising the at least one additional material; and stacking a plurality of said layers to form the stack.

7. The method of Claim 6, further comprising sintering said plurality of layers after said compressing.

8. A method for producing an edge seal for a stack of a plurality of layers comprising a catalyst material, said method comprising:

mixing a catalyst material with at least one additional material to form a mixture; and,

during a reaction, converting the at least one additional material to a gas-impermeable material forming an edge seal.

9. A method for producing an edge seal for a stack of a plurality of layers, each layer comprising a catalyst material, said method comprising:

compressing the catalyst material, thereby forming a layer;

stacking a plurality of layers to form the stack; and

applying a gas-impermeable and temperature-resistant material to said stack, thereby forming an edge seal.

10. The method of Claim 9, said applying is by immersing, plasma spraying, or flame spraying.

11. The method of Claim 9, further comprising
applying a soldering foil on the stack; and
exposing said stack to an elevated temperature
wherein the soldering foil penetrates into the layers and
forms a gas-tight edge seal.

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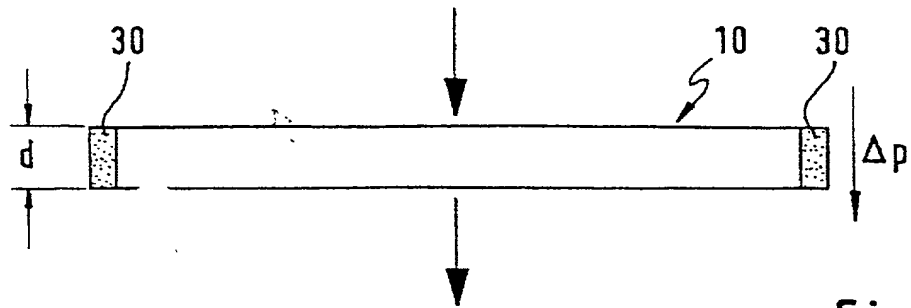


Fig. 1

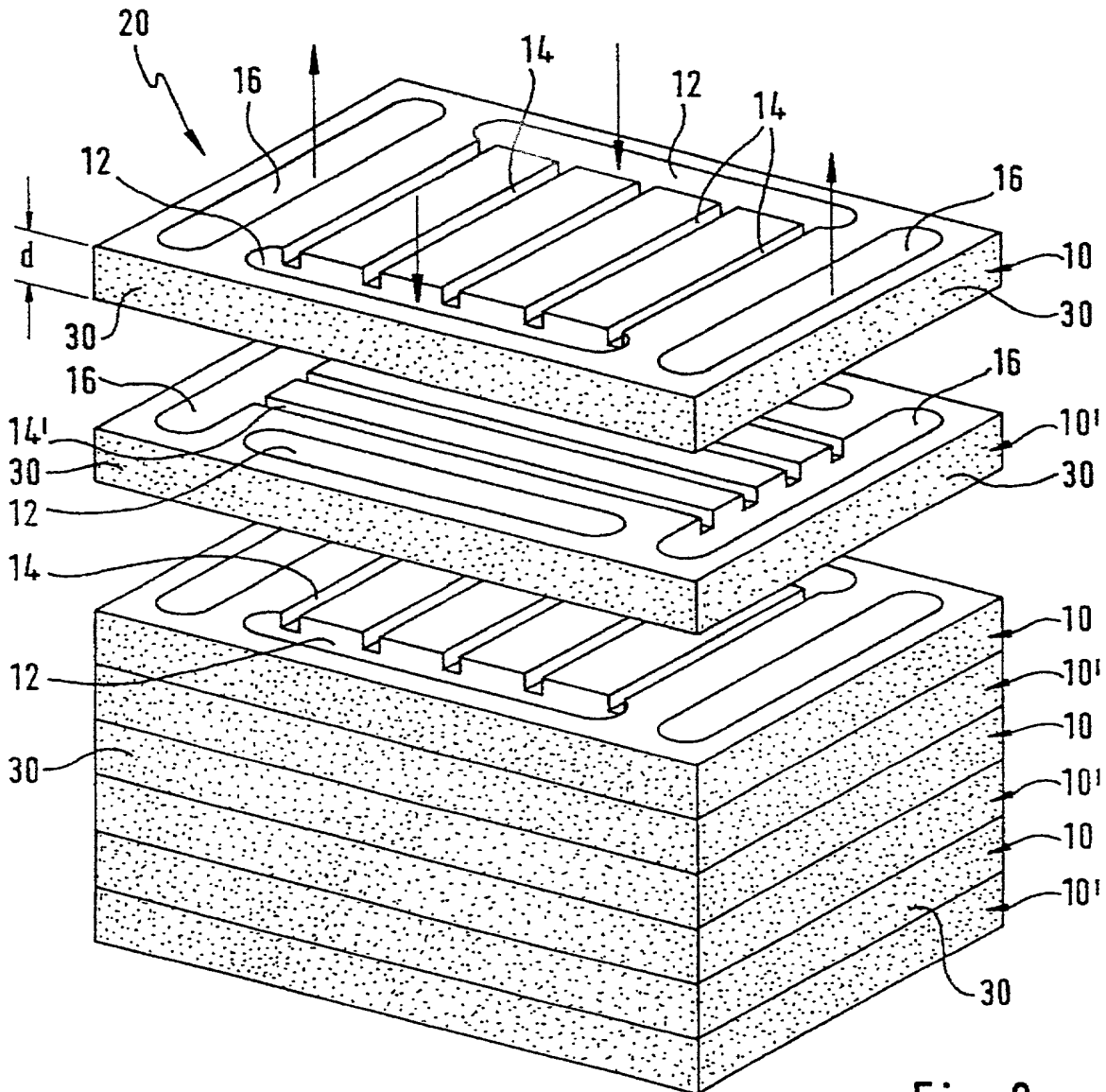
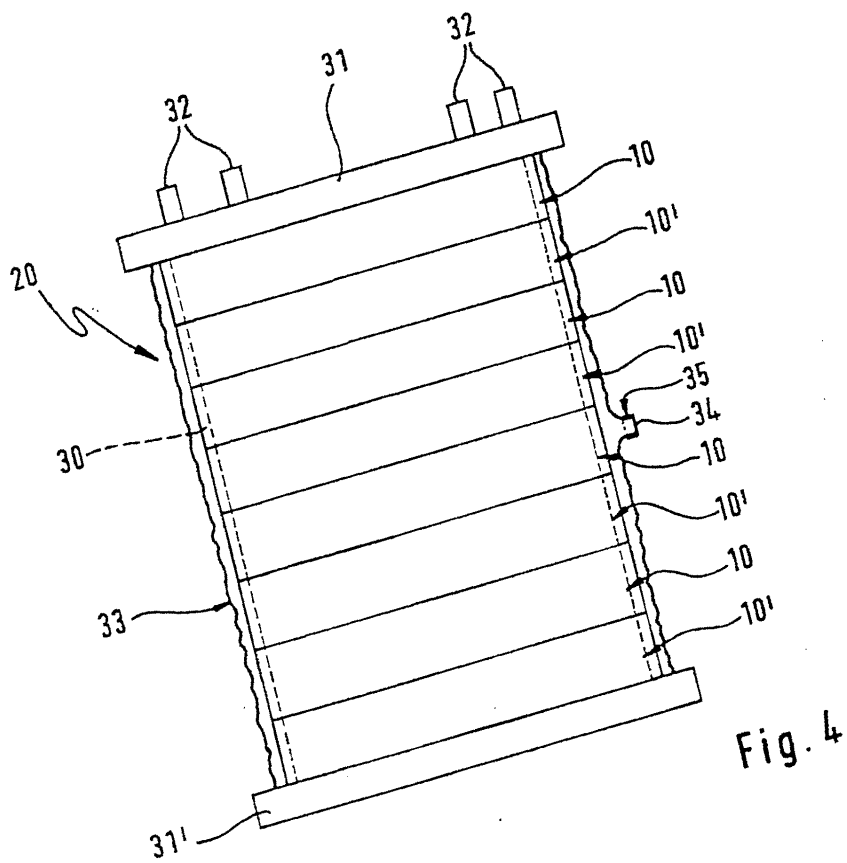
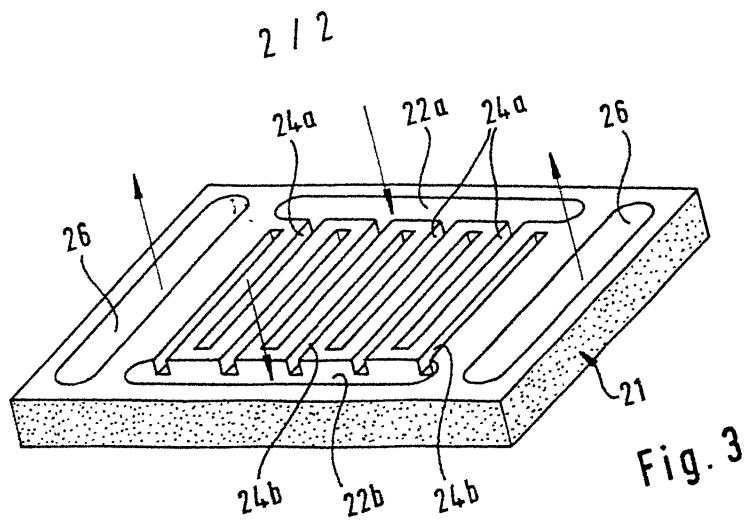


Fig. 2

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DECLARATION AND POWER OF ATTORNEY - PATENT APPLICATION

As a below named inventor, I hereby declare that my citizenship, postal address and residence are as stated below; that I verily believe I am the original, first and sole inventor (if only one inventor is named below) or a joint inventor (if plural inventors are named below) of the invention entitled:

AN APPARATUS FOR A CARRYING OUT A HETEROGENEOUSLY CATALYZED REACTION AND A METHOD FOR PRODUCING SAME

the specification of which

X is attached hereto, or
 _____ was filed on _____ as Application Serial No. _____ and
 was amended on _____ (if applicable).

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above. I acknowledge the duty to disclose all information known to be material to patentability as defined in 37 CFR §1.56. I hereby claim foreign priority benefits under Title 35, United States Code §119 of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed:

Prior Foreign Application(s)			Priority Claimed
<u>199 44 185.5</u> (Number)	<u>Germany</u> (Country)	<u>15 September, 1999</u> (Day/Month/Year)	<u>Yes</u>
_____ (Number)	_____ (Country)	_____ (Day/Month/Year)	_____

I hereby claim the benefit under Title 35, United States Code, §120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code, §112, I acknowledge the duty to disclose all information known to be material to patentability as defined in 37 CFR §1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application:

(Application Serial No.)	(Filing Date)	(Status)
_____	_____	_____

I hereby appoint as principal attorneys Herbert I. Cantor, Reg. No. 24,392; James F. McKeown, Reg. No. 25,406; Donald D. Evenson, Reg. No. 26,160; Joseph D. Evans, Reg. No. 26,269; Gary R. Edwards, Reg. No. 31,824; Jeffrey D. Sanok, Reg. No. 32,169; and Richard R. Diefendorf, Reg. No. 32,390, to prosecute and transact all business in the Patent and Trademark Office connected with this application and any related United States and international applications. Please direct all communications to:

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DECLARATION AND POWER OF ATTORNEY

Attorney Docket No. 1748X/49133

Page 2

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under §1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

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